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中华人民共和国国家知识产权局

邮政编码: 100037 北京市阜成门外大街 2 号万通新世界广场 8 层 中国国际贸易促进委员会专利商标事务所 陈昕 CO/3269	发文日期
申请号: 02102099X	
申请人: 株式会社日本触媒	
发明创造名称: 固体床多管式反应器及其使用方法	

第一次审查意见通知书

1. 应申请人提出的实审请求,根据专利法第 35 条第 1 款的规定,国家知识产权局对上述发明专利申请进行实质审查。

根据专利法第 36 条第 2 款的规定,国家知识产权局决定自行对上述发明专利申请进行审查。

2. 申请人要求以其在:

JP 专利局的申请日 2001 年 01 月 25 日为优先权日,
专利局的申请日 年 月 日为优先权日,
专利局的申请日 年 月 日为优先权日,
专利局的申请日 年 月 日为优先权日,
专利局的申请日 年 月 日为优先权日。

申请人已经提交了经原申请国受理机关证明的第一次提出的在先申请文件的副本。

申请人尚未提交经原申请国受理机关证明的第一次提出的在先申请文件的副本,根据专利法第 30 条的规定视为未提出优先权要求。

3. 经审查,申请人:

年 月 日提交的 不符合实施细则第 51 条的规定;
年 月 日提交的 不符合专利法第 33 条的规定;
年 月 日提交的

4. 审查针对的申请文件:

原始申请文件。 审查是针对下述申请文件的

申请日提交的原始申请文件的权利要求第 项、说明书第 页、附图第 页;
年 月 日提交的权利要求第 项、说明书第 页、附图第 页;
年 月 日提交的权利要求第 项、说明书第 页、附图第 页;
年 月 日提交的权利要求第 项、说明书第 页、附图第 页;
年 月 日提交的说明书摘要, 年 月 日提交的摘要附图

5. 本通知书是在未进行检索的情况下作出的。

本通知书是在进行了检索的情况下作出的。

本通知书引用下述对比文献(其编号在今后的审查过程中继续沿用):

编号 文件号或名称 公开日期 (或抵触申请的申请日)

1 CN 1049368 C 2000.02.16
2 CN 1003353 B 1989.02.22

6. 审查的结论性意见:

关于说明书:

申请号 02102099X

 申请的内容属于专利法第 5 条规定的不授予专利权的范围。 说明书不符合专利法第 26 条第 3 款的规定。 说明书不符合专利法第 33 条的规定。 说明书的撰写不符合实施细则第 18 条的规定。 关于权利要求书： 权利要求 _____ 不具备专利法第 22 条第 2 款规定的创造性。 权利要求 1-7 不具备专利法第 22 条第 3 款规定的创造性。 权利要求 _____ 不具备专利法第 22 条第 4 款规定的实用性。 权利要求 _____ 属于专利法第 25 条规定的不授予专利权的范围。 权利要求 _____ 不符合专利法第 26 条第 4 款的规定。 权利要求 _____ 不符合专利法第 31 条第 1 款的规定。 权利要求 _____ 不符合专利法第 33 条的规定。 权利要求 _____ 不符合专利法实施细则第 2 条第 1 款关于发明的定义。 权利要求 _____ 不符合专利法实施细则第 13 条第 1 款的规定。 权利要求 _____ 不符合专利法实施细则第 20 条的规定。 权利要求 1, 4, 6 不符合专利法实施细则第 21 条的规定。 权利要求 _____ 不符合专利法实施细则第 22 条的规定。 权利要求 _____ 不符合专利法实施细则第 23 条的规定。

上述结论性意见的具体分析见本通知书的正文部分。

7. 基于上述结论性意见，审查员认为：

 申请人应按照通知书正文部分提出的要求，对申请文件进行修改。 申请人应在意见陈述书中论述其专利申请可以被授予专利权的理由，并对通知书正文部分中指出的不符合规定之处进行修改，否则将不能授予专利权。 专利申请中没有可以被授予专利权的实质性内容，如果申请人没有陈述理由或者陈述理由不充分，其申请将被驳回。

8. 申请人应注意下述事项：

(1) 根据专利法第 37 条的规定，申请人应在收到本通知书之日起的三个月内陈述意见，如果申请人无正当理由逾期不答复，其申请将被视为撤回。

(2) 申请人对其申请的修改应符合专利法第 33 条的规定，修改文本应一式两份，其格式应符合审查指南的有关规定。

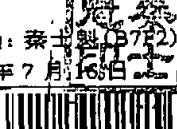
(3) 申请人的意见陈述书和/或修改文本应邮寄或递交国家知识产权局专利局受理处，凡未邮寄或递交给受理处的文件不具备法律效力。

(4) 未经预约，申请人和/或代理人不得前来国家知识产权局专利局与审查员举行会晤。

9. 本通知书正文部分共有 3 页，并附有下述附件：

 引用的对比文件的复印件共 2 份 21 页。 审查员：秦士魁 (B7F2)
2004 年 7 月 16 日

审查部门 材料审查部

21301
2003 A回函请寄：100088 北京市海淀区蓟门桥西土城路 6 号 国家知识产权局专利局受理处收
(注：凡寄给审查员个人的信函不具有法律效力)

PAGE 6/50 * RCVD AT 10/13/2004 12:59:28 PM [Eastern Daylight Time] * SVR:USPTO-EXRF-1/1 * DNIS:8729310 * CSID:651 699 7901 * DURATION (mm:ss):13:52

中华人民共和国国家知识产权局

第一次审查意见通知书正文

申请号：02102099X

审查员：秦士魁
代码：B7F2

本申请涉及一种【固定床多管式反应器及其使用方法】，如说明书所述，本申请要解决的技术问题是“【提供一种在填充催化剂固体粒状物用于制造各种物质时，能够长期稳定地制造目的产物的固定床反应器及其使用方法（说明书1页17-18行；审查员：上述译文中“各种物质”和“目的产物”是否是“同一种物质”？该段译文不清楚，请申请人斟酌）】”。经审查，现提出如下的审查意见。

1. 权利要求1的主题类型不清楚，不符合实施细则第20条第1款的规定，权利要求1的前序部分的发明主题是 固定床多管式反应器，而特征部分 …在固定床…反应管中。”涉及的是方法特征。
2. 独立权利要求1缺少达到发明目的的必要技术特征（审查员：该方法特征缺少达到发明目的的必要技术特征，即实现“在各反应管中填充的催化剂的量或压力降是均一的”具体手段）。不符合实施细则第21条第2款的规定。
3. 权利要求1不具备创造性，不符合专利法第22条第3款的规定。对比文件1公开了一种“固定床反应器催化剂的装卸方法”，并具体公开了以下的技术特征“采用由吸粒管（1）…组成的装填催化剂系统…”（参见对比文件1的全文）；对比文件2公开了一种“在列管式固定床反应器中进行反应的方法及装置”，并具体公开了以下的技术特征“在固定床反应器列

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管间进行气固流化换热的固体颗粒为球形颗粒..." (参见对比文件2的全文)。由此可见, 该权利要求区别于对比文件1和对比文件2的特征仅在于本发明申请明确提出"在各反应管中填充的催化剂的量或压力降是均匀的" (审查员: 没有实现该目的的具体手段), 而采取对比文件1和2具体手段也可实现上述的目的 (审查员: "在各反应管中填充的催化剂的量或压力降是均匀的"这一要求是固定床反应器的基本要求, 对本领域的技术人员是常识问题)。这区别仅仅是所属技术领域的公知常识。对比文件1和对比文件2技术领域相同, 在对比文件1的基础上结合对比文件2, 并结合所属技术领域中的公知常识, 得出该权利要求所要求保护的技术方案, 对所属技术领域的技术人员来说是显而易见的, 而且它们的结合没有产生预料不到的技术效果, 因此该权利要求不具备突出的实质性特点和显著的进步, 因而不具备创造性。

4. 权利要求2-7之任1的权利要求均存在上述1或2的问题, 同时其与对比文件1和2的特征比较, 也不具备创造性, 不符合专利法第22条第3款的规定。

5. 从属权利要求 4, 6 本身是一个多项从属权利要求, 它引用了在前的多项从属权利要求 3 等, 因此不符合实施细则第 23 条第 2 款的规定。申请人应当对该权利要求的引用关系进行修改。

6. 如果申请人同意上述1和2的意见, 则应修改发明名称。

7. 请申请人再校对部分们现申请文本, 译文最好吻合中文的表达习惯。

申请人应当在本通知书指定的答复期限内对本通知书提出的问题逐一进

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行答复,必要时应进行修改专利申请文件,否则本申请将难以获得批准.申请人对申请文件的修改应当符合专利法第33条的规定,不得超出原说明书和权利要求书记载的范围.

申请人的修改文件应当包括:第一,修改部分的原文复印件,采用红色钢笔在该复印件上标注出所作的增加,删除或替换;第二,重新打印的替换页(一式二份),用于替换相应的原文.申请人应当确保上述二部分在内容上的一致性.

(人)

[19]中华人民共和国国家知识产权局

[51]Int. Cl.

B01J 8/06

[12]发明专利说明书

[21]ZL 专利号 95107485.7

[45]授权公告日 2000年2月16日

[11]授权公告号 CN 1049368C

[22]申请日 1995.7.14 [24]授权日 1999.10.9

[74]专利代理机构 石油化工科学研究院专利事务所
代理人 周定秀

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审查员 姚杰

权利要求书2页 说明书8页 附图页数3页

[54]发明名称 固定床反应器催化剂的装卸方法

[57]摘要

本发明描述了直立多管固定床反应器催化剂的装、卸方法及其主要设备。本发明所述装、卸催化剂的方法是指采用由吸粒管(1)为主要部件组成的装、卸催化剂系统,装填和卸出直径≤15mm的催化剂或填料颗粒的过程。其主要设备吸粒管(1)的入口端有三个圆周各间隔120°的平台,用来疏通反应管(3)内催化剂颗粒架桥,清除催化剂颗粒粘结、挂壁和堵塞现象。本发明具有装、卸催化剂系统结构简单、催化剂颗粒架桥、阻塞易清除、操作方便等优点,也适用于在生产非正常中断时,需要在惰性气体保护催化剂条件下装、卸催化剂。

ISSN1008-4274

专利文献出版社出版

权利要求书

1. 一种直立多管固定床反应器中的催化剂的卸出方法, 其特征在于采用由吸粒管(1)为主要部件组成的卸催化剂系统, 按照下面的操作步骤卸出催化剂: 将吸粒管(1)的入口端穿过同心气流入出三通(2)插入反应管(3)中, 直抵催化剂的床层界面, 打开与同心气流入出三通(2)相连的阀门(11), 启动真空抽气装置(8), 由(8)产生的负压气流, 通过反应管(3)与吸粒管(1)之间的环形空间至催化剂床层界面, 调节抽气气流速度, 使负压气流夹带着催化剂颗粒经吸粒软管(4)入气固分离器(5), 过细的催化剂粉尘经气固分离器(5)顶部的抽气口逸出, 再经粉尘过滤器(7)捕集间歇取出, 催化剂颗粒收集于气固分离器(5)的底部, 打开(5)底部的阀(10)和催化剂容器(6)上方的阀(12), 催化剂经排粒管(9)间歇排放至催化剂容器(6)中。

2. 根据权利要求1所说的卸出催化剂方法, 其特征在于在惰性气体保护下卸催化剂, 在卸催化剂系统中, 与同心气流入出三通(2)相连的阀(11)直接与惰性气体源相连。

3. 根据权利要求1所说的卸出催化剂方法, 其特征在于, 催化剂粒度 $<15\text{mm}$, 也装卸同样粒度的惰性填料。

4. 根据权利要求1所说的卸出催化剂方法, 其特征在于, 在卸催化剂系统中, 每个部件之间, 是通过橡胶部件密封或管线连接。

5. 根据权利要求1所说的卸出催化剂方法, 其特征在于, 吸粒管(1)的管径 db 小于反应管(3)的管径(Dra), 其长度比反应管(3)长5-10%, 吸粒管(1)的入口端有三个圆周各间隔 120° 的平齿, 平齿的高度(h)约为吸粒管(1)直径 db 的三分之一, 吸粒管(1)直径 db 与催化剂直径 dp 之比为 $db/dp > 3$ 。

6. 一种直立多管固定床反应器催化剂的装填方法, 其特征在于采用由吸粒管(1)为主要部件组成的装填催化剂系统, 按照下面的操作步骤装填催化剂: 打开吸粒软管(4)与催化剂装填漏斗(14)之间的阀(16), 关闭(14)下面的阀门(17)和(14)侧上方的阀门(18)以及与(14)的上方、同心气流入出三通(2)、粉尘过滤器(7)相连的阀门(19), 将吸粒管(1)插入装有催化剂的容器(6)中, 直抵催化剂界面, 打开(6)上方的阀门(12), 启动真空抽气装置(8), 调节抽气气流速度, 使(8)产生的负压气流夹带着催化剂颗粒经吸粒管(1)和吸粒软管(4)进入催化剂装填漏斗(14), 过细的催化剂粉尘从

(14)的顶部抽气口逸出,经粉尘过滤器(7)捕集间歇取出,当催化剂装填漏斗(14)中的催化剂的数量达到预定值时,停止抽气,打开阀门(18、19),使系统恢复常压,关闭阀门(16),打开阀门(17),催化剂装填漏斗(14)中的催化剂,经装剂管(15)穿过同心气流入出三通(2)缓慢地放入反应管(3)中。

7. 根据权利要求 3 所说的装填催化剂的方法, 其特征在于在惰性气体保护下装填催化剂, 在装填催化剂系统中催化剂容器(6)上方的阀(12)、催化剂装填漏斗(14)侧上方的阀(18)直接与惰性气体源相连。

8. 根据权利要求 3 所说的装填催化剂方法, 其特征在于, 催化剂粒度 $< 15\text{mm}$, 也装填同样粒度的惰性填料。

9. 根据权利要求 3 所说的装填催化剂方法, 其特征在于, 在装填催化剂系统中, 每个部件之间, 是通过橡胶部件密封或管线连接。

10. 根据权利要求 3 所说的装填催化剂方法, 其特征在于, 吸粒管(1)的管径 d_b 小于反应管(3)的管径(D_{ra}), 其长度比反应管(3)长 5-10%, 吸粒管(1)的入口端有三个圆周各间隔 120° 的平齿, 平齿的高度(h)约为吸粒管(1)直径 d_b 的三分之一, 吸粒管(1)直径 d_b 与催化剂直径 d_p 之比为 $d_b/d_p \geq 3$ 。

说 明 书

固定床反应器催化剂的装卸方法

本发明涉及一种固定床反应器催化剂的装卸方法，确切地说是关于直立多管固定床反应器中催化剂的装填和卸出方法及其主要设备。

颗粒的气流输送，已广为各有关工业、农业、交通运输部门应用，但在化工系统中，应用于直立多管固定床反应器中催化剂的装填及卸出报导较少。直立多管反应器内的催化剂卸出，多采用下卸出方法，即在反应器底部卸催化剂口处放出，这种卸催化剂方法常常因反应管直径较小，反应管内颗粒架桥、粘结堵塞、或与管壁粘附，导致催化剂卸出不尽，难以顺利操作，尤其在需要对催化剂进行惰性气体保护及需要进行筛分或分离催化剂与填料颗粒时，由于设备庞大，粉尘污染严重，给操作人员带来更大的麻烦。R079865公开了一种筒形反应器中催化剂连续卸出的设备及方法，该法是采用两只交替操作的旋风分离器及真空抽气方法连续进行操作，卸出筒形反应器内的废催化剂，但未涉及直立多管反应器，也未涉及装填及卸出催化剂时容易架桥、堵塞等问题。Br75-06251公开了一种直立多管反应器催化剂卸出装置及其改进方法，该专利提出，将一只可移动管头接在反应管出口，穿过此管头插入一只长矛器具至反应管内，真空抽气，使反应管内床层催化剂颗粒流动起来，强制疏通架桥，清除堵塞，达到顺利卸出催化剂的目的，为了使反应管内整个床层催化剂流动起来，需要空气流量大，抽气设备大，能耗高，操作也不方便。

本发明的目的是提供一种直立多管固定床反应器催化剂的卸出方法。

本发明的另一目的是提供一种直立多管固定床反应器催化剂的

装填方法。

本发明的第三个目的是提供一种用于装填或卸出催化剂系统中的主要部件—吸粒管。

本发明是通过下述方案实现的：

当卸出催化剂时，由真空抽气装置产生的负压气流，通过反应管与吸粒管之间的环形空间至催化剂床层界面，夹带着催化剂颗粒，经吸粒管进入气固分离器沉降收集于其下部，然后间歇取出；过细的催化剂粉尘则随负压气流经粉尘过滤器加以捕集，尾气由真空系统排出。

当装填催化剂时，由真空抽气装置产生的负压气流，通过吸粒管将催化剂颗粒从催化剂容器中夹带至催化剂装填漏斗中，在催化剂达到预定数量后，停止抽气，打开催化剂装填漏斗下部的开关，催化剂即经装剂管放入反应管中。

用于装、卸催化剂系统中的主要部件吸粒管，其入口端有三个圆周各间隔120°的平齿，用来疏通反应管内催化剂颗粒架桥，清除颗粒粘结，挂壁和堵塞现象。

下面结合附图详细描述本发明直立多管固定床反应器卸出和装填催化剂的方法以及用于装填或卸出催化剂系统中主要部件吸粒管(1)的结构。

图1为本发明卸出催化剂颗粒的流程示意图。

图2为本发明装填催化剂颗粒的流程示意图。

图3为吸粒管(1)入口端剖析图。

由图1所示，卸出催化剂系统是由下述部件和设备构成：将可移动和旋转的金属吸粒管(1)的入口端穿过连接反应管(3)入口端的同心

气流入出三通(2)，直抵反应管(3)中的催化剂床层界面，吸粒管(1)的另一端通过吸粒软管(4)与气固分离器(5)连接，(5)的顶部与粉尘过滤器(7)相连，(5)的底部装有阀(10)，通过排粒管(9)与催化剂容器(6)相连。粉尘过滤器(7)与真空抽气装置(8)相连，阀11与同心气流入出三通(2)相连。

本发明卸出催化剂的操作步骤如下：将吸粒管(1)的入口端穿过同心气流入出三通(2)插入反应管(3)中，直抵催化剂的床层界面，打开与同心气流入出三通(2)相连的阀门(11)，启动真空抽气装置(8)，由(8)产生的负压气流，通过反应管(3)与吸粒管(1)之间的环形空间至催化剂床层界面，在床层界面处上下移动吸粒管或调节负压气流速度，负压气流夹带着催化剂颗粒经吸粒软管(4)入气固分离器(5)并收集于其底部，过细的催化剂粉尘经气固分离器(5)顶部的抽气口逸出，再经粉尘过滤器(7)捕集，从过滤器(7)处间歇取出，打开(5)底部的阀(10)和催化剂容器(6)上方的阀(12)，催化剂经排粒管(9)间歇排放至催化剂容器(6)中。

如果需要在惰性气体保护条件下卸出催化剂，在卸催化剂系统中，可将阀(11)直接与惰性气体源(如氮气)相连。

由图2所示，装填催化剂系统由下述部件和设备构成：将吸粒管(1)的入口端插入催化剂容器(6)并伸至催化剂界面，另一端通过吸粒软管(4)与催化剂装填漏斗(14)相连，装填漏斗(14)下部通过装剂管(15)穿过同心气流入出三通(2)插入反应管(3)中，装填漏斗(14)的上部与粉尘过滤器(7)以及同心气流入出三通(2)连接，粉尘过滤器(7)与真空抽气装置(8)相通。

本发明装填催化剂的具体操作步骤是：打开吸粒软管(4)与催化

剂装填漏斗(14)之间的阀门(16),关闭(14)下面的阀门(17)和(14)侧上方的阀门(18)以及与(14)上方、同心气流入出三通(2)以及粉尘过滤器(7)相连的阀门(19),将吸粒管(1)插入装有催化剂的容器(6)中,直抵催化剂界面,打开(6)上方的阀门(12),启动真空抽气装置(8),在床层界面上下左右移动吸粒管头或调节抽气气流速度,使(8)产生的负压气流夹带着催化剂颗粒经吸粒管(1)和吸粒软管(4)进入催化剂装填漏斗(14),过细的催化剂粉尘从(14)的顶部抽气口逸出,经粉尘过滤器(7)捕集间歇取出,当催化剂装填漏斗(14)中的催化剂数量达到预定值时,停止抽气,打开阀门(18、19),使系统恢复正常压,关闭阀门(16),打开阀门(17),催化剂装填漏斗(14)中的催化剂,经装剂管(15)穿过同心气流入出三通(2)缓慢地放入反应管(3)中。

如果需要在惰性气体保护条件下装填催化剂,在装填催化剂系统中,可将催化剂容器(6)上方的阀(12),催化剂装填漏斗(14)侧上方的阀(18)直接与惰性气体源(如氮气)相连。

本发明所说的装、卸方法中,催化剂粒度≤15mm,也可以装卸同样粒度的惰性填料。

本发明所说的装、卸方法中,装卸催化剂系统每个部件之间是通过橡胶部件密封或管线连接。

由图3所示,吸粒管(1)入口端有三个圆周各间隔120°的平齿,平齿的高度(h)约为吸粒管(1)的直径(d_b)的三分之一(即h=1/3d_b)。

吸粒管(1)是用于装填或卸出催化剂系统中的关键部件,吸粒管(1)的管径(d_b)小于反应管(3)的管径(D_{re}),其长度大于反应管(3)长度约5-10%。吸粒管(1)通常是由多只较短的金属管螺纹连接而成,

可以上下、左右移动、正反方向旋转，其入口端有三个圆周各间隔 120° 的平齿，用来疏通反应管内催化剂颗粒架桥，清除颗粒粘结、挂壁和堵塞现象，随着催化剂的卸出，床层界面下降，吸粒管(1)也随之下移，以保证连续地将反应管(3)内的催化剂全部卸出。吸粒管(1)的另一端通过吸粒软管(4)与气固分离器(5)相连，起着引导气固输送流的作用。吸粒管(1)直径 d_b 与催化剂或填料颗粒直径 d_p 之比为 $d_b/d_p > 3$ 。

本发明所说的同心气流入出三通(2)，不仅可以用于与大气相通条件下装填或卸出催化剂，也适用于在惰性气体(如氮气)保护条件下装填或卸出催化剂或填料颗粒。

本发明所说的气固分离器(5)，用于卸催化剂系统中，其侧面有气固流入口，下端有固体颗粒收集部及排放口，上部为扩大段、下部为锥形体的收缩段，负压气流或惰性气流进入气固分离器(5)扩大段后，气流速度骤减，使得催化剂或填料颗粒不能被夹带，而落入分离器(5)底部，达到气固分离之目的，也可以采用旋风分离结构进行气固分离。

本发明所说的催化剂装填漏斗(14)是一个标记有容量刻度的，透明的或局部透明的，可见内部催化剂界面的气固分离器，用于装填催化剂系统中，其顶部有气流引出口，和惰性气体或大气输入口，侧面上端有气固流入口，下端有固体颗粒收集段及排放口，以利于气固分离及确定每次反应管(3)内装填催化剂或填料颗粒的数量。

本发明所说的催化剂容器(6)的上方有一个惰性气体或大气的入出口以及阀(12)，对于卸催化剂系统，在容器(6)的下部还有一个排放催化剂的阀(13)，当容器(6)中的催化剂或填料颗粒装满时，打开

阀(13)，将催化剂或填料颗粒排放到专用容器中。

本发明所说的排粒管(9)可以随意上下、左右移动，在卸催化剂系统中，若将排粒管(9)与吸粒管(1)交换位置，可以将卸催化剂系统转变为装填催化剂系统。

本发明也可以对多个反应管的催化剂或填料颗粒同时装填或卸出，以加快装、卸催化剂的过程。

本发明具有以下优点。

1. 装、卸催化剂系统结构简单，不需其它辅助设备，效率高。
2. 由于在装、卸催化剂系统中，使用了关键设备—吸粒管，因此易于装填及卸出各种容易架桥、粘结、挂壁、堵塞的催化剂及填料颗粒。
3. 所用真空抽气装置负荷小，设备小，能耗低。由于直立多管固定床反应器中的反应管的直径通常都较小，只需很小的气体流量，就能产生足够高的气流线速，达到夹带催化剂或填料颗粒的要求。同时，由于本发明使用了吸粒管，仅仅要求在吸粒管入口端附近的负压气流达到夹带催化剂颗粒的要求，因此，所需压力降很低，对真空抽气设备要求抽气负荷低，抽气量小，抽气设备小，而已有技术Br75-06251所述方法要求床层催化剂流动起来，所需压力降高，气体流量大，能耗高，而且还需辅助长茅器具清除架桥和堵塞现象。
4. 适用于装卸较大尺寸的颗粒材料，如直径 $d_p=8\sim15\text{mm}$ 的催化剂，以及惰性填料，如瓷球、刚玉球等均可顺利地装入及卸出，而不致架桥。
5. 使用灵活，功能多，适用于反应管细而长的多管直立反应器装卸催化剂，特别适用于在生产非正常中断时，需要在惰性气体保护

催化剂条件下处理反应器运行中发生各种故障时使用。

6. 由于全部操作均在负压下进行, 排出气流都经过粉尘过滤器, 因此, 有毒粉尘污染小。

下面的实施例将对本发明作进一步说明

实例 1

一直立多管反应器, 壳体直径 $D_a=800\text{mm}$, 其中有144根直径 $D_{ra}=38 \times 3.0\text{mm}$ 、长3400mm的反应管, 按照预定装量的要求, 每根反应管先装入床高300mm、直径为10mm的刚玉球, 然后再装入床高2800mm、直径 $d_p=1.5-2.5\text{mm}$ 的球粒催化剂, 再装入床高300mm直径为5mm的刚玉球, 直到全部装完。起初用手工倒入直径为10mm的刚玉球时, 在反应管内发生架桥阻塞现象的有十余根, 未能达到预定高度要求。采用本发明所述卸催化剂方法, 按照图1所示装置流程, 将吸粒管伸入堵塞点, 启动抽气系统, 将架桥阻塞的刚玉球吸出, 然后再按照图2所示装置流程, 进行刚玉球和催化剂的装填, 使催化剂的装填工作顺利完成。所用吸粒管是由两只长1800mm, 外径 $d_s=30\text{mm}$ 的金属管以及连接管头组装而成。

实例 2

在同实例1的直立多管反应器内, 采用本发明所述方法, 按照图1所示的装置流程卸废催化剂和填料颗粒。将吸粒管伸入到反应管中, 直抵床层界面, 启动抽真空系统, 首先吸出 $d_p=5\text{mm}$ 的瓷球, 并收集于气固分离器中, 当瓷球吸尽时, 停止抽气, 将瓷球排放到容器(6)中, 然后收集到专用容器中, 下次待用。调节抽气气速, 将吸粒管继续插

入至废催化剂界面,此时 $d_p=1.5-2.5\text{mm}$ 的废催化剂顺利卸出,未出现任何架桥阻塞现象,当废催化剂吸尽时,停止抽气,将废催化剂排放 在容器(6)中 ,然后收集在废催化剂专用容器中,最后,按照同样步骤,将 $d_p=10\text{mm}$ 的瓷球全部卸出,如此重复,逐一卸出144根反应管中的催化剂和填料颗粒,所有的催化剂粉尘,捕集于布袋过滤器中。

实例 3

一个由56根反应管组成的直立多管反应器,每根反应管的直径 $D_{\text{ext}}=38 \times 3.0\text{mm}$,长3400mm,先按照图2所示装置流程装填催化剂和填料颗粒,进行胺化反应,反应完后,采用8只双排三角排列的吸粒管组合装置卸出废催化剂和填料颗粒,先卸出300mm高度的装在上层的填料颗粒($d_p=5.0\text{mm}$),然后卸出中段2800mm高度的催化剂颗粒($d_p=2.0\text{mm}$),最后卸出下层300mm高度的直径为10mm的填料颗粒、采用同实例1的抽真空系统,将废催化剂和填料颗粒全部卸出,使卸出过程大大加快。

说 明 书 附 图

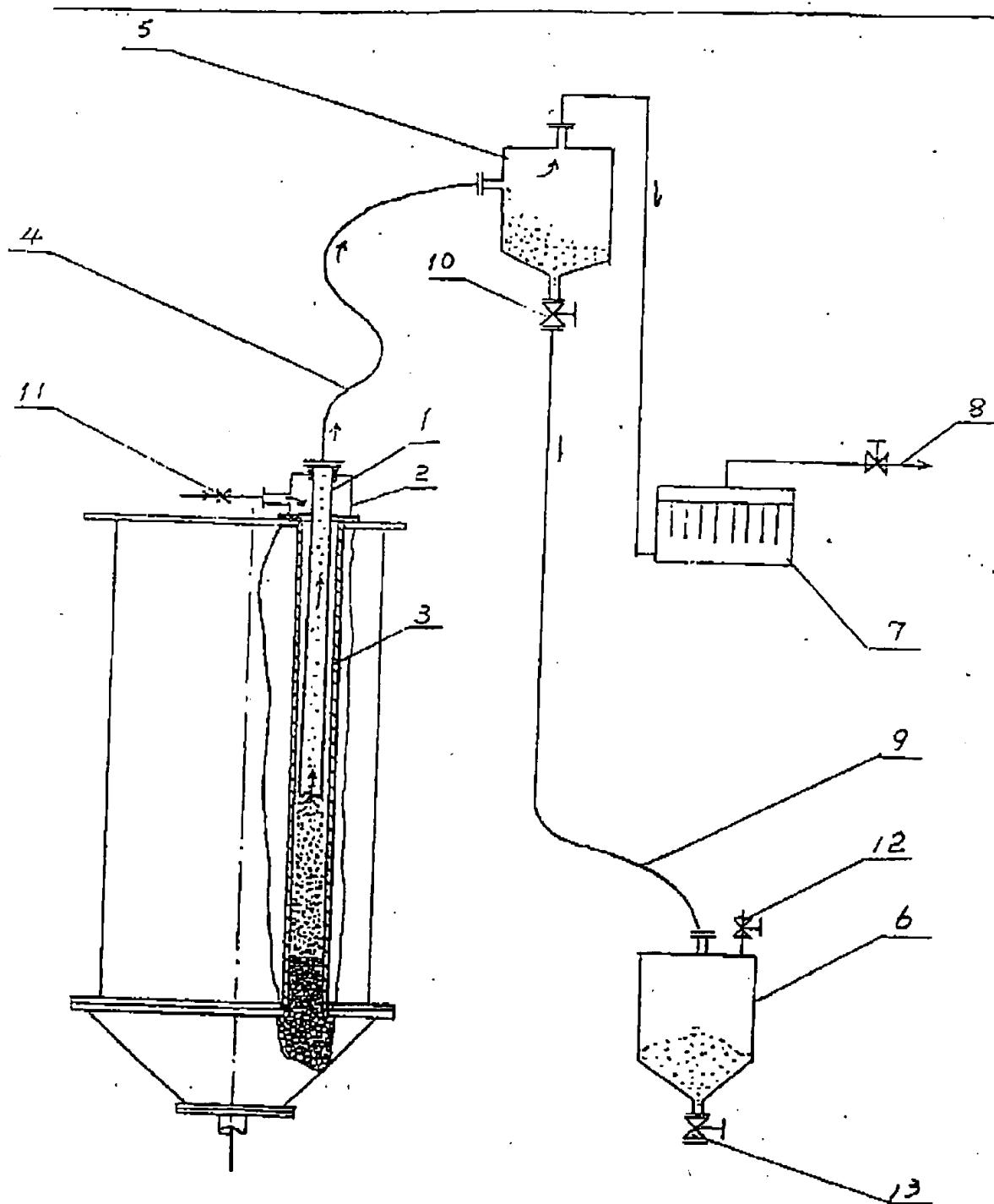


图 1

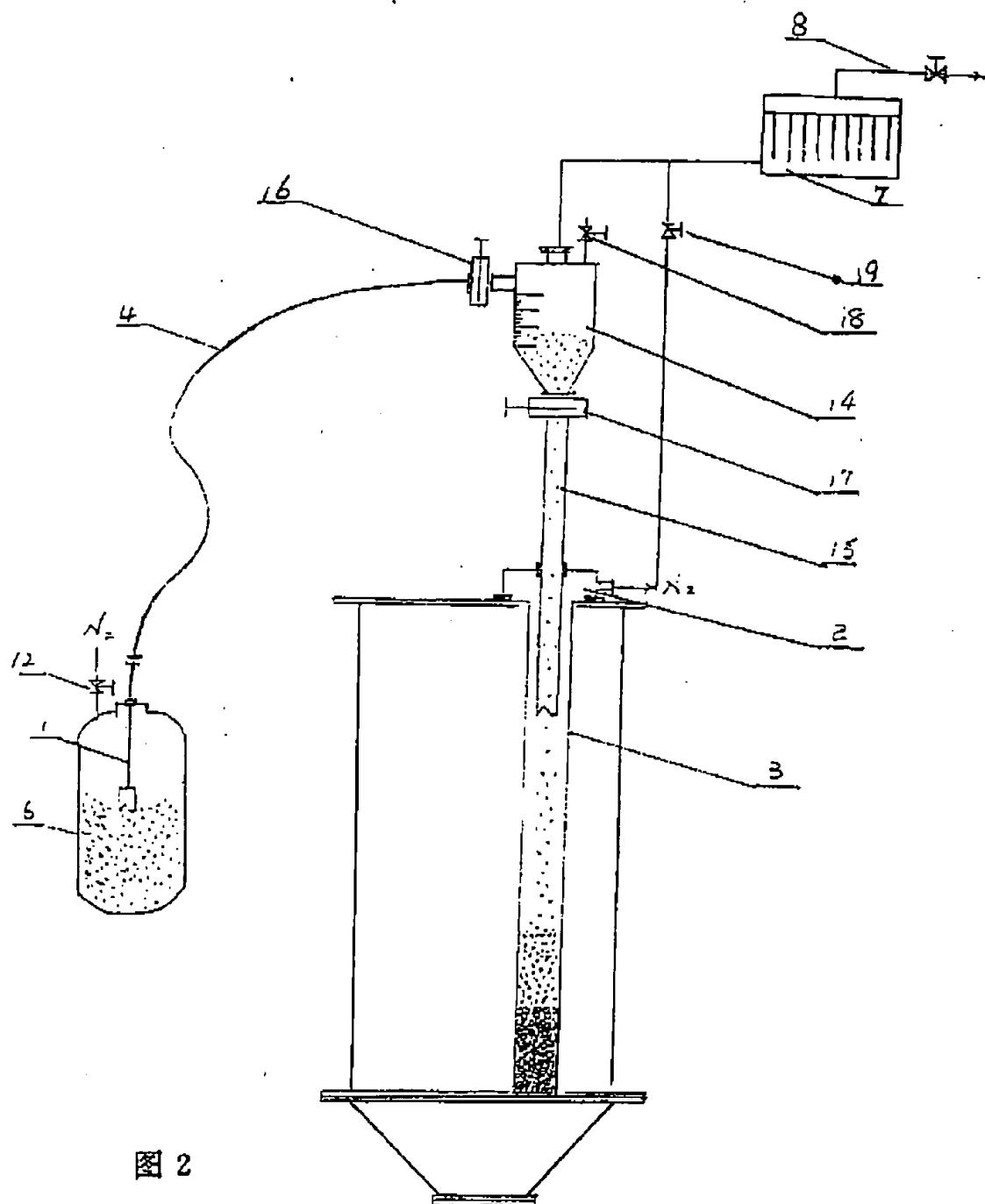


图 2

- 2 -

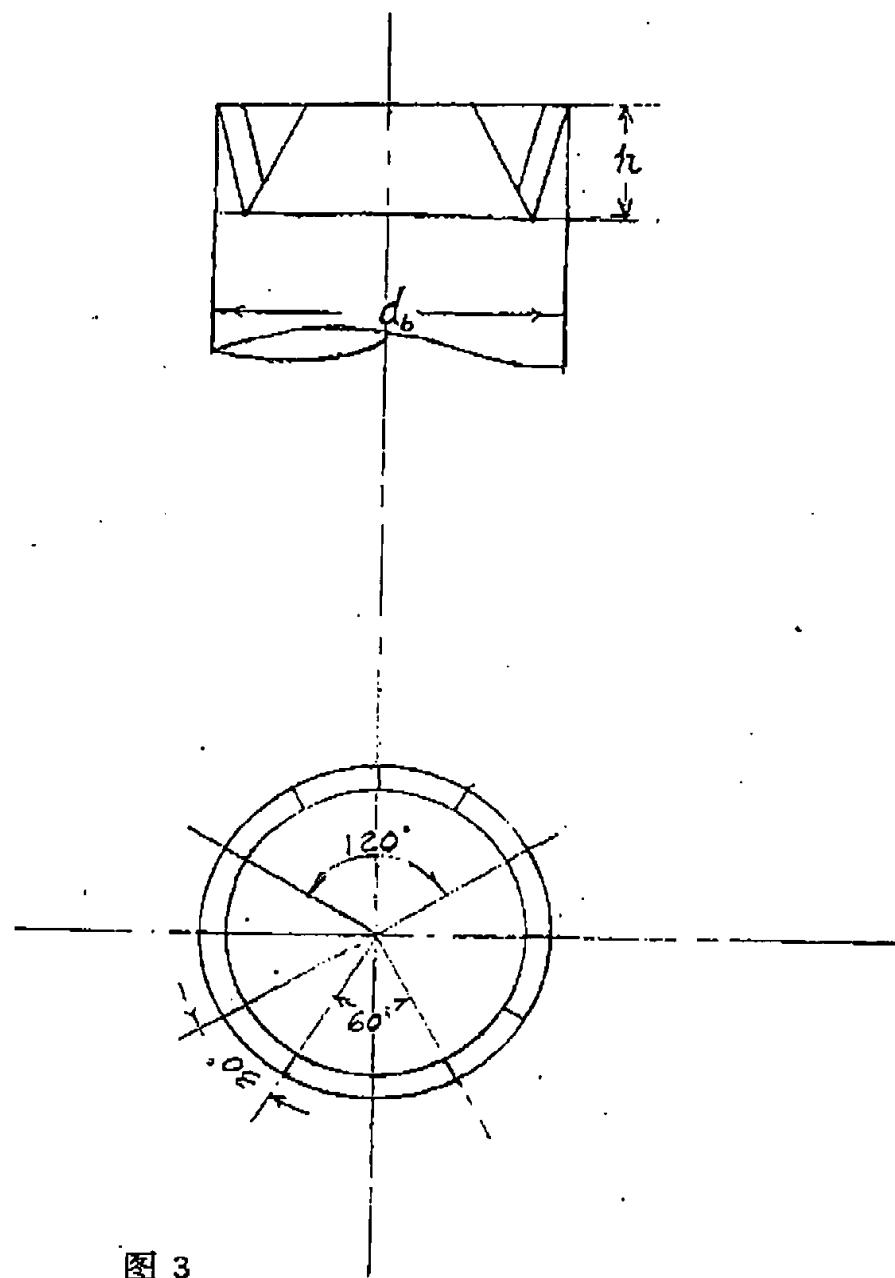


图 3

— 3 —

(b)

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Method for Loading and Unloading Fixed Bed Reactor Catalyst

The invention describes a method and apparatus for loading and unloading vertical and multi-tube fixed bed reactor catalyst. The method for loading and unloading catalyst according to the invention is meant a course to load and unload a catalyst or a filler particle with a diameter ≤ 15 mm using a loading and unloading system consisting of particulate absorbing tube (1) as the main element. The inlet end of the main apparatus particulate absorbing tube (1) has three flat gears of each circle spacing 120° , which are used to dredge the bridge formation of particle catalyst in reactor tubes (3) and eliminates the adhesion, lodge and block phenomenon of particle catalyst. The invention has advantages of simple system structure of loading and unloading catalyst, easy elimination of bridge and block formation of particle catalyst, convenient operation, and is also suitable for loading and unloading catalyst in inert atmosphere when the production is interrupted non-normally.

Method for Loading and Unloading Fixed Bed Reactor Catalyst

The invention relates to a method for loading and unloading a fixed bed reactor catalyst, particularly to a method for loading and unloading a vertical and multi-tube fixed bed reactor catalyst and main apparatuses therefor.

The gas-flow conveying of particles is widely used in industry, agriculture and traffic departments, but in chemical field, its use in loading and unloading catalyst in vertical and multi-tube fixed bed reactor has been rarely reported. The catalyst in the vertical and multi-tube reaction is usually unloaded using lower unloading method, that is, the catalyst is discharged from the outlet at the bottom of the reactor. This method of unloading catalyst makes the discharge of catalyst incomplete and the operation of difficult due to small diameter of reactor tubes, bridge formation, lodge and block, or adhesion on tube wall of particle catalyst in reactor tubes. Specifically when the catalyst is required to be protected in inert atmosphere and to be sieved or the catalyst is separated from filler particles, due to bulky equipments and serious dust pollution, larger troubles are brought to the operators. RO79865 discloses an apparatus and method for continuous unloading a catalyst in cylinder reactor. The method comprises discharging the spent catalyst in the cylinder reactor by continuous operation of two alternatively operating cyclones and vacuum air pump, but it does not relate to vertical and multi-tube reactors, nor bridge and block easily formed when loading and unloading catalysts. Br 75-06251 discloses an apparatus and improved method for unloading a vertical and multi-tube reactor catalyst. The patent raises to connect a movable tube head to an outlet of the reactor tube, and insert a lance through the tube head into the reactor tube, vacuum air pump to make the particle catalyst in bed layer of the reactor tube flow to forcibly dredge bridge formation and eliminate block so as to smoothly discharge the catalyst. In order to make the whole bed layer

catalyst in the reactor tube flow, large air flow and bulky pumping equipments are required, and thus energy consumption is high and operation is inconvenient.

The purpose of the invention is to provide a method for unloading a vertical and multi-tube fixed bed reactor catalyst.

Another purpose of the invention is to provide a method for loading a vertical and multi-tube fixed bed reactor catalyst.

The third purpose of the invention is to provide main element for use in a system of loading or unloading catalyst, i.e. particulate absorbing tube.

The invention is achieved by following solutions:

When a catalyst is unloaded, gas flow of negative pressure generated by a vacuum air pumping apparatus passes through annular space between the reactor tube and the particulate absorbing tube to catalyst bed layer interface, carries the particle catalyst through the particulate absorbing tube into gas-solid separator to settle and collected at the lower portion of the separator, and then is discharged out intermittently; superfine catalyst dust passes through dust collector together with gas flow of negative pressure and is collected, tail gas is exhausted by a vacuum system.

When a catalyst is loaded, gas flow of negative pressure generated by a vacuum air pumping apparatus passes through the particulate absorbing tube to carry the particle catalyst from the catalyst container to catalyst loading funnel, after the catalyst reaches to a certain amount, air pumping is stopped, the switch at the lower portion of the catalyst loading funnel is opened, then the catalyst is introduced into the reactor tube through a reagent loading tube.

Regarding the main element particulate absorbing tube for use in a system of loading and unloading catalyst, its inlet end has three flat gears of each circle spacing 120°, which are used to dredge the bridge formation of particle catalyst in reactor tubes and eliminate the adhesion, lodge and block phenomenon of particle catalyst.

The method for loading and unloading a vertical and multi-tube fixed bed reactor catalyst and the structure of the main apparatus particulate absorbing tube for use in the system of loading and unloading catalyst according to the invention are described detailedly in combination with figures as follows:

Fig. 1 is a sketch map of flow of unloading particle catalyst according to the invention.

Fig. 2 is a sketch map of flow of loading particle catalyst according to the invention.

Fig. 3 is an explosion view of inlet end of particulate absorbing tube (1).

It can be seen from Fig. 1, catalyst unloading system is composed of following elements and apparatus: the inlet end of movable and rotary metal particulate absorbing tube (1) passes through concentric gas flow inlet outlet three-way pipe (2) to catalyst bed layer interface in reactor tube (3), the other end of particulate absorbing tube (1) is connected to gas-solid separator (5) through a particulate absorbing tube (4), the top of the separator (5) is connected to dust collector (7), the bottom of the separator (5) is mounted with a valve (10) and connected to a catalyst container (6) through particulate discharging tube (9). The dust collector (7) is connected to a vacuum air pumping apparatus (8), a valve (11) is connected to the concentric gas flow inlet outlet three-way pipe (2).

The operation steps of unloading catalyst according to the invention are as

follows: inserting the inlet end of particulate absorbing tube (1) through concentric gas flow inlet outlet three-way pipe (2) into reactor tube (3) until bed layer interface of catalyst, opening a valve (11) connected to concentric gas flow inlet outlet three-way pipe (2), starting up vacuum air pumping apparatus (8), passing the gas flow of negative pressure produced by (8) through the annular space between the reactor tube (3) and particulate absorbing tube (1) to catalyst bed layer interface, moving the particulate absorbing tube up and down at bed layer interface or adjusting negative pressure gas flow rate, the negative pressure gas flow carrying the particle catalyst through particulate absorbing tube (4) into gas-solid separator (5) and being collected at its bottom, superfine catalyst dust exhausting from the outlet at the top of gas-solid separator (5), collecting it through a dust collector (7) and discharging it intermittently from the collector (7), opening a valve (10) at the bottom of (5) and a valve (12) over the catalyst container (6), discharging the catalyst intermittently through particulate discharging tube (9) into a catalyst container (6).

If the catalyst is required to be discharged under protection conditions of inert gases, in the system for unloading catalyst, a valve (11) may be directly connected to inert gas source (such as nitrogen).

It can be seen from Fig. 2, the catalyst loading system is composed of the following elements and apparatus: an inlet end of particulate absorbing tube (1) is inserted into a catalyst container (6) and extended to catalyst interface, the other end is connected to a catalyst loading funnel (14) through a particulate absorbing tube (4), the lower portion of loading funnel (14) passes through reagent loading tube (15) and concentric gas flow inlet outlet three-way pipe (2) and then is inserted into reactor tube (3), the upper portion of loading funnel (14) is connected to a dust collector (7) and concentric gas flow inlet outlet three-way pipe (2), dust collector (7) is connected to a vacuum air pumping

apparatus (8).

The operations steps of loading catalyst according to the invention are as follows: opening a valve (16) between a particulate absorbing tube (4) and a catalyst loading funnel (14), closing a valve (17) under (14), a valve (18) side-over (14) and a valve (19) connected to the upper of (14), concentric gas flow inlet outlet three-way pipe (2) and a dust collector (7), Inserting a particulate absorbing tube (1) into a container (6) filled with catalyst, straight to the catalyst interface, opening the valve (12) over (6), starting up the vacuum air pumping apparatus (8), moving particulate absorbing head around the bed layer interface or adjusting air pumping gas flow rate to make negative pressure of gas flow produced by (8) carry the particle catalyst through a particulate absorbing tube (1) and a particulate absorbing tube (4) into catalyst loading funnel (14), superfine catalyst dust is exhausted from the outlet at the top of (14), collected through a dust collector (7) and discharged intermittently, when the amount of catalyst in catalyst loading funnel (14) reaches to a predetermined value, air pumping is stopped, valves (18, 19) are opened to reconvert the system to normal pressure, a valve (16) is closed, a valve (17) is opened, the catalyst in catalyst loading funnel (14) is slowly placed through reagent loading tube (15) and concentric gas flow inlet outlet three-way pipe (2) into the reactor tube (3).

If the catalyst is required to be loaded under the protection conditions of inert gases, in the system of loading catalyst, a valve (12) over the catalyst container (6) and a valve (18) side-over the catalyst loading funnel (14) may be directly connected to inert gas source (such as nitrogen).

In the method of loading and unloading according to the invention, the particle size of catalyst is ≤15mm, inert filler with same particle size may be loaded and unloaded.

In the method of loading and unloading according to the invention, each element of catalyst loading and unloading system is sealed with rubber elements or connected with pipeline.

It can be seen from Fig. 3, the inlet end of particulate absorbing tube (1) has three flat gears of each circle spacing 120°, the height (h) of flat gears is about 1/3 of the diameter (d_b) of particulate absorbing tube (1) (i.e. $h=1/3 d_b$).

Particulate absorbing tube (1) is used to the key element in the system of loading or unloading catalyst, the diameter (d_b) of particulate absorbing tube (1) is less than the tubular diameter (D_m) of the reactor tube (3), its length is about 5-10% greater than that of the reactor tube (3). Particulate absorbing tube (1) is usually made by screw connecting a plurality of short metal pipes, and can move up and down, right and left, and rotate in positive and negative directions, its inlet end has three flat gears of each circle spacing 120°, which are used to dredge the bridge formation of particle catalyst in reactor tubes and eliminate the adhesion, ledge and block phenomenon of particle catalyst. With the unloading of catalyst, bed layer interface is lowered, particulate absorbing tube (1) moves down to ensure continuous and complete unloading of catalyst in the reactor tube (3). The other end of particulate absorbing tube (1) is connected to gas-solid separator (5) through particulate absorbing tube (4), acting on leading gas-solid conveying stream. The ratio of the diameter (d_b) of particulate absorbing tube (1) to the diameter (d_p) of catalyst or filler particles is $d_b/d_p \geq 3$.

The concentric gas flow inlet outlet three-way pipe (2) according to the invention can be not only used for loading or unloading catalyst at a atmosphere-communicating condition but also used for loading or unloading catalyst or filler particles at the protection condition of inert gases (such as

nitrogen).

The gas-solid separator (5) according to the invention is used in the system of unloading catalyst, its side has an inlet of gas solid stream, its lower end has a collection portion of solid particles and discharge outlet, its portion is broadening section, its lower portion is conical contracting section, after negative pressure gas flow or inert gas flow enters into the broadening section of gas-solid separator (5), gas flow rate is reduced abruptly so that the catalyst or filler particles cannot be carried and fall into the bottom of separator (5) so as to achieve separation of the gas and solid. Gas-solid separation may be carried out using cyclone separation structure.

The catalyst loading funnel (14) according to the invention is a gas solid separator with a marked capacity scale, which is transient or locally transient, and its inner catalyst interface can be seen. It is used in the system of loading catalyst. Its top has an outlet for gas flow, an inlet of inert gas or free air, the upper end of its side has an inlet of gas solid stream, its lower end has solid particle collection section and a discharge outlet, to be beneficial to separation of gas and solid and determine the amount of catalyst or filler particles to be loaded in the reactor tube (3) every time.

There are an inlet or outlet of inert gas or free air and a valve (12) above the catalyst container (6) according to the invention. For a system for unloading catalyst, there is also a valve (13) for discharging catalyst at the lower portion of the container (6). When the catalyst or filler particles in the container (6) are filled up, the valve (13) is opened, the catalyst or filler particles are discharged into a special container.

The particulate discharging tube (9) according to the invention may be optionally moveable up and down, right and left. In the system of unloading

catalyst, if the positions of particulate discharging tube (9) and particulate absorbing tube (1) are exchanged, the catalyst unloading system can be converted into the catalyst loading system.

The invention can simultaneously load or unload the catalyst or filler particles in a plurality of reactor tubes so as to expedite the loading and unloading of catalyst.

The invention has the following advantages:

1. The structure of catalyst loading and unloading system is simple, other assistant equipments are not needed, and efficiency is high.
2. Since the key apparatus, particulate absorbing tube, is used in the catalyst loading and unloading system, the loading and unloading of various catalyst and filler particles which are easily bridged, bond, lodged or blocked can be easily carried out.
3. The used vacuum air pumping apparatus is small in load and equipment, and low in energy. Since the diameter of reactor tube in vertical and multi-tube fixed bed reactor is usually small, very small flow amount of gas can produce enough high linear velocity of gas flow so as to achieve the requirement of carrying catalyst or filler particles. At the same time, since the invention uses particulate absorbing tubes, it is only required that the gas flow of negative pressure round the inlet end of the particulate absorbing tube fulfills the requirement of carrying catalyst particles, required pressure drop is very low, vacuum air pumping apparatus is required to have low load of air pump, small air pumping amount and small air pumping apparatus, however, said method of the prior art Br75-06251 requires bed layer catalyst movable, required pressure drop is high, flow amount of gas is large, and energy consumption is high, and assistant lances are required to eliminate bridge and block formation.
4. It is suitable for loading and unloading particulate materials of large size, for

example, catalysts of diameter $d_p=8-15$ mm as well as inert fillers, such as ceramic balls, corundum balls, can be loaded and unloaded smoothly, and bridge cannot be formed.

5. Use is flexible, and function is multiple. It is suitable for loading and unloading catalyst in vertical and multi-tube reactor of thin and long reactor tubes, specifically suitable in treatment of various exceptions occurred during the circulation of reactors when the production is interrupted non-normally and the catalyst is required to be protected with inert gases.
6. Since all operations are carried out at negative pressure, exhausted gas flow passes through dust collector, the pollution of toxic dust is small.

Following examples further describe the invention.

Example 1

A vertical and multi-tube reactor of diameter $D_o=800$ nm, in which 144 reactor tubes of $D_{ra}=38 \times 3.0$ mm and length of 3400 mm are contained, according to predetermined loading requirement, each reactor tube is loaded with corundum balls with bed height of 300mm and diameter of 10mm, and spherical catalysts with bed height of 2800mm and diameter $d_p = 1.5-2.5$ mm, and then corundum balls with bed height of 300mm and diameter of 5mm. At first, when the corundum balls of diameter of 10mm are poured in manually, bridge and block formations occur in about ten reactor tubes, and predetermined height requirements are not achieved. By using the method of unloading catalyst according to the invention, and according to the flow of apparatus shown in Fig. 1, a particulate absorbing tube is stretched into the block point, and air pumping system is started up to suck bridged and blocked corundum balls out, then according to apparatus flow shown in Fig. 2, corundum balls and catalysts are loaded so as to complete the loading of catalysts smoothly. Used particulate absorbing tube is assembled with two metal tubes of length 1800mm and outer diameter $d_b=30$ mm and a connection

tube head.

Example 2

In vertical and multi-tube reactor identical to In example 1, spent catalyst and filler particles are unloaded by using the method of invention and according to the apparatus flow shown in Fig. 1. A particulate absorbing tube is stretched into the reactor tube until the bed layer interface, vacuum air pumping system is started up, ceramic balls of $d_p=5$ mm, and collected into gas-solid separator, when the ceramic balls are sucked out, air pumping is stopped, the ceramic balls are discharged into the container (6), and then collected into a special container, stand-by. Air pumping rate is adjusted, the particulate absorbing tube is continuously inserted until the interface of spent catalyst, herein the spent catalyst of $d_p=1.5-2.5$ mm is discharged smoothly. No bridge and block are found. When the spent catalyst is sucked out, air pumping is stopped, the spent catalyst is discharged into the container (6), and then collected into a special container. At last, according to same steps, ceramic balls of $d_p=10$ mm are discharged completely, repeatedly, the catalyst and filler particles in 144 reactor tubes are unloaded one by one, all catalyst dust is collected into cloth envelop collector.

Example 3

A vertical and multi-tube reactor consisting of 56 reactor tubes, each reactor tube has a diameter $D_{in}=38\times 3.0$ mm, and a length of 3400mm, catalyst and filler particles are loaded according to the apparatus flow shown in Fig. 2 and subjected to amination reaction, upon completion, spent catalyst and filler particles are unloaded using 8 particulate absorbing tubes assembly in doublebank triangle arrangement, the filler particles ($d_p=6.0$ mm) of 300 mm height mounted at upper layer are first discharged, then the catalyst particles ($d_p=2.0$ mm) of 2800 mm height at middle section are discharged, and last the filler particles (diameter 10mm) of 300mm height at lower layer are discharged.

By using the same vacuum air pumping system as in example 1, the spent catalyst and filler particles all discharged so as to expedite the unloading greatly.

Claims

1. A method for unloading catalyst in vertical and multi-tube fixed bed reactor, characterized in that the catalyst is unloaded using a catalyst unloading system with a particulate absorbing tube (1) as main element and according to the following steps: inserting the inlet end of particulate absorbing tube (1) through concentric gas flow inlet outlet three-way pipe (2) into reactor tube (3) until bed layer interface of catalyst, opening a valve (11) connected to concentric gas flow inlet outlet three-way pipe (2), starting up a vacuum air pumping apparatus (8), passing the gas flow of negative pressure produced by (8) through the annular space between the reactor tube (3) and particulate absorbing tube (1) to catalyst bed layer interface, adjusting air pumping gas flow rate to make the negative pressure gas flow carry particle catalyst through a particulate absorbing tube (4) into a gas-solid separator (5), exhausting superfine catalyst dust from the outlet at the top of the gas-solid separator (5) and discharging the dust from a dust collector (7) intermittently, collecting the particle catalyst at the bottom of the gas-solid separator (5), opening a valve at the bottom of (5) and a valve (12) over a catalyst container (6), discharging the catalyst intermittently through particulate discharging tube (9) into the catalyst container (6).
2. The method for unloading catalyst according to claim 1, characterized in that the catalyst is unloaded under the protection of an inert gas, in the catalyst unloading system, a valve (11) connected to concentric gas flow inlet outlet three-way pipe (2) is directly connected to an inert gas source.
3. The method for unloading catalyst according to claim 1, characterized in that the catalyst has a particle size ≤ 15 mm, an inert filler having same particle size is also unloaded.

4. The method for unloading catalyst according to claim 1, characterized in that in catalyst unloading system, each element is sealed with a rubber element or connected by pipeline.
5. The method for unloading catalyst according to claim 1, characterized in that the tube diameter db of particulate absorbing tube (1) is less than the tube diameter (Dra) of a reactor tube (3), and its length is 5-10% longer than that of the reactor tube (3), the inlet end of particulate absorbing tube (1) has three flat gears of each circle spacing 120° , the height (h) of the flat gears is about $1/3$ of the diameter db of particulate absorbing tube (1), the ratio of the diameter db of particulate absorbing tube (1) to that of catalyst dp is $db/dp \geq 3$.
6. A method for loading catalyst in vertical and multi-tube fixed bed reactor, characterized in that the catalyst is loaded using a catalyst unloading system with a particulate absorbing tube (1) as main element and according to the following steps: opening a valve (16) between a particulate absorbing tube (4) and a catalyst loading funnel (14), closing a valve (17) under (14), a valve (18) side-over (14) and a valve (19) connected to the upper of (14), concentric gas flow inlet outlet three-way pipe (2) and a dust collector (7), inserting a particulate absorbing tube (1) into a container (8) filled with catalyst until the catalyst interface, opening the valve (12) over (6), starting up the vacuum air pumping apparatus (8), adjusting air pumping gas flow rate to make negative pressure of gas flow produced by (8) carry the particle catalyst through a particulate absorbing tube (1) and a particulate absorbing tube (4) into catalyst loading funnel (14), superfine catalyst dust is exhausted from the outlet at the top of (14), collected through a dust collector (7) and discharged intermittently, when the amount of catalyst in catalyst loading funnel (14) reaches to a predetermined value,

air pumping is stopped, valves (18, 19) are opened to reconvert the system to normal pressure, a valve (16) is closed, a valve (17) is opened, the catalyst in catalyst loading funnel (14) is slowly introduced through reagent loading tube (15) and concentric gas flow inlet outlet three-way pipe (2) into the reactor tube (3).

7. The method for loading catalyst according to claim 3, characterized in that the catalyst is loaded under the protection of an inert gas, in the catalyst loading system, a valve (12) over the catalyst container (6) and a valve (19) side-over a catalyst loading funnel (14) are directly connected to the inert gas source.
8. The method for loading catalyst according to claim 3, characterized in that the catalyst has a particle size ≤ 15 mm, an inert filler having same particle size is also loaded.
9. The method for loading catalyst according to claim 3, characterized in that in catalyst loading system, each element is sealed with a rubber element or connected by pipeline.
10. The method for loading catalyst according to claim 3, characterized in that the tube diameter db of particulate absorbing tube (1) is less than the tube diameter (Dra) of a reactor tube (3), and its length is 5-10% longer than that of the reactor tube (3), the inlet end of particulate absorbing tube (1) has three flat gears of each circle spacing 120° , the height (h) of the flat gears is about $1/3$ of the diameter db of particulate absorbing tube (1), the ratio of the diameter db of particulate absorbing tube (1) to that of catalyst dp is $db/dp \geq 3$.